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DEVELOPMENT OF A NUCLEONIC PARTICULATE EMISSION GAUGE

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FINAL REPORT

ENVIRONMENTAL PROTECTION AGENCY

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16. Abstracts

A project was conducted to design, fabricate, and test a particulate emission monitor utilizing a beta radiation attenuation and filter sampler technique. The instrument was to have a minimum sensitivity of 125 ± 25 micrograms per cubic meter and be capable of operation in either a manual or automatic mode of operation. Carbon-14 was selected as the optimum isotope and a filter "cassette" was used to collect the particles. The effect of moisture absorption by various types of filter media were investigated and reported. A series of tests were performed on automobiles using leaded, low-leaded and non-leaded fuels. The results of these tests are reported as well as recommendations for future effort.

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FINAL REPORT

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Environmental Protection Agency
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Contract No. 68-02-0210

25 February 1972

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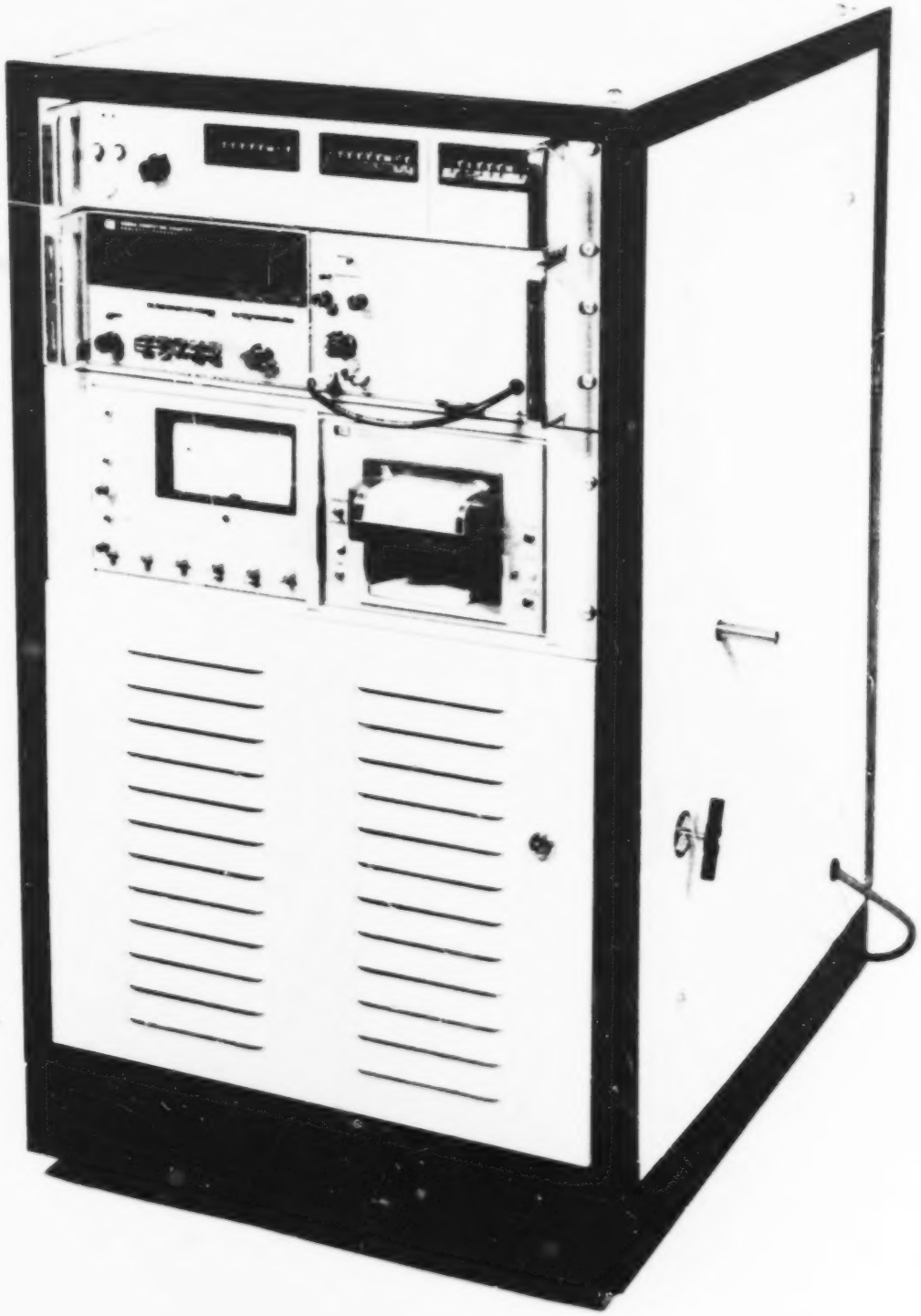
1.0 INTRODUCTION

This report is the final report to Environmental Protection Agency Contract No. 68-02-0210. Upon acceptance by the EPA, this report represents completion of contractor-required delivered items.

In June of 1971, the EPA contracted with Industrial Nucleonics (IN) to develop a beta gauge and filter sampler for measurement of particulate emissions from automobile exhaust. The gauge was to have a minimum sensitivity of 125 micrograms per cubic meter ($\mu g/m^3$) with no specified upper limit. Further, the filter collection system was to have an efficiency using a dioctyl phthalate aerosol of at least 99.9%. A gauge to meet these requirements has been designed, built, tested, and delivered and is as shown in Figure 1-1.

A Hewlett-Packard Computing Counter has been used to perform the calculations and switching functions. In addition, a Fischer and Porter Swirlmeter has been provided for reliable determination of gas flow. The inclusion of these two units allows high accuracy and repeatability with respect to computation. This was deemed necessary in order to fulfill the wide variety of applications against which the unit was to be tested. The major thrust of the program has been to design a research tool rather than a prototype so that evaluation of the unit could be made on the basis of the suitability of beta gauging for the various applications, not merely IN's ability to produce such a gauge. Certainly, for many specific applications, a considerable reduction in complexity and cost is possible.



VEHICLE EXHAUST PARTICULATE EMISSION GAUGE

Figure 1-1

As part of the contract, this unit was tested recently at one of the EPA's Cincinnati facilities and at the Dow Chemical Company in Midland, Michigan. The Cincinnati series was primarily intended for unit de-bug and checkout. The tests at Dow were performed on automobiles using leaded (3cc/gal tetraethyl lead), low-leaded (0.5 cc/gal TEL) and non-leaded (0.0 cc/gal TEL) fuels. Good results were obtained using leaded and low-leaded fuels when compared to gravimetric measurements. However, less desirable results were obtained with no-leaded fuels. No conclusion may be drawn from the non-leaded tests since correlation between two different size (4 inch and 2 inch filters) gravimetric measurements could not be achieved, thereby leaving no standard against which to compare the gauge.

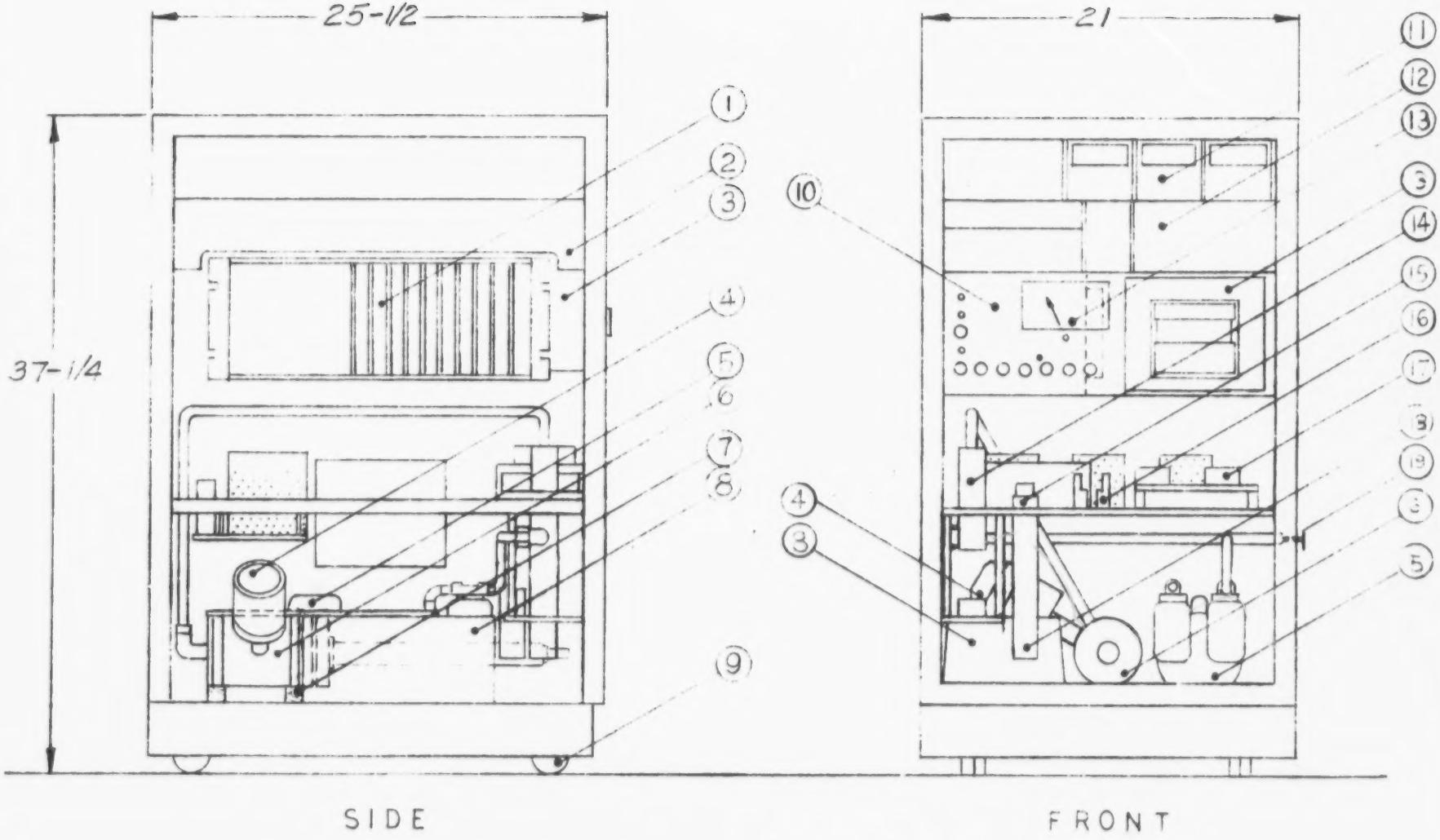
The details of these tests, and the details surrounding IN's selection of this particular design constitute the text of this report. While reference will occasionally be made to operation of the unit, the reader is referred to the Operation Manual accompanying the unit for specific operation, maintenance and safety procedures to be followed.

1.1 Principal Components of the Gauge

This gauge has been designed primarily as a research tool. There has been little attention devoted to minimizing the size and weight and maximizing the portability as efforts of this type were considered to be beyond the scope of this contract. The device has been fitted

with wheels, however, to assist in moving the unit from place to place. Also the Computing Counter and Systems Programmer are quite easily removed (See Operation Manual) for further ease of transport. A complete list of the principal components and their model numbers is as follows (See Figure 1-2):

1. Electronic circuitry (printed circuit boards). Access to this circuitry is obtained by removing the side panel
2. Welded cabinet brace
3. Printer. Hewlett-Packard Model No. 5055A
4. Flowmeter preamplifier
5. Vacuum pump. GAST Model No. 0822
6. Flowmeter. Fischer and Porter "Swirlmeter"
7. Flowmeter supports
8. Flowmeter electronics
9. Swivel caster (four identical double-wheel units)
10. Control panel with switches and indicator lights
11. Systems programmer, Hewlett-Packard Model No. 5376A
12. Computing counter, Hewlett-Packard Model No. 5360A
13. Flow rate display meter
14. Particulate Collection station. Station at which gas is drawn through the filter and particles are collected
15. Nucleonic source enclosure and filter locating mechanism
16. Magazine to hold 20 filters



CABINET AND PRINCIPAL COMPONENTS
(SIDE PANEL AND FRONT DOOR REMOVED)

Figure 1-2

17. Filter transporting mechanism actuator, Cliftronics R28S1 32 rotary solenoids

18. Radiation detector housing. Station at which filter and collected particulate masses are detected.

19. Flow regulating valve.

1.2 Specifications

This gauge has been overdesigned for many specific applications. This is a direct result of emphasis being placed on accuracy over a wide range and flexibility. The beta counting times and pumping times may seem astronomical and unnecessary. The magnitude of these numbers is, however, an added bonus received by inclusion of the HP Computing Counter and Systems Programmer. It is also correct to say that the computational errors are infinitesimal by comparison to the errors in the sensing heads (flowmeter and beta counting). This is as it should be for an instrument of this nature. The limitation of the gauge is contained solely in the limitations of the sensors used, namely the state-of-the-art in stabilizing photomultiplier tubes, the state-of-the-art in measuring absolute flow, and process repeatibility. This necessarily implies that improvement in any one of these three areas will result in improved measurement accuracy automatically.

The major specifications follow, but some prefacing comments are necessary. First, it is difficult to state a range of the gauge in terms of stream concentration unless some limit is placed on both flow rate and time. This is a result of there being both a minimum and a maximum

total mass which can be detected with the current design. The minimum and maximum mass determinations are stated with the assumptions on flow rate and time clarifying the stream concentration.

Second, probably the greatest limiting factor on accuracy at high concentrations is the ability to measure the total sample volume correctly. The Swirlmeter measures volumetric flow and therefore to achieve a measurement in terms of an "Actual Cubic Foot", use must be made of the suggested correction factor in the Operations Manual. For most applications where this factor is not included, the flow accuracy is ± 2%, excluding corrections to "Standard Cubic Foot."

1.2.1 Major Specifications (assumes zero process error)

	Specification	Value	Remarks
1.	Effective Filter Area	3 cm^2	
2.	Flow Rate	20-120 l/min	
3.	Minimum Concentration	80 ± 16 $\mu g/m^3$ (2σ)	
4.	Maximum Concentration	3×10^6 ± 2% $\mu g/m^3$ (assumes no flow-meter correction)	120 l/min for 20 min
5.	Beta Counting Range	10^{-6} sec to 2×10^{11} sec.	actual
6.	Beta Counting Range	30 sec. to 300 sec.	practical
7.	Pumping Time	10^{-6} sec to 2×10^{11} sec.	actual
8.	Pumping Time	30 sec to 3×10^4 sec.	practical
9.	Minimum Mass Resolution	± 60 μg (2σ)	

10.	Maximum Mass Determination	30,000 ± 600 μg (2σ)	
11.	Flow Absolute Accuracy	± 0.75% ACFM	Manufacturer's specification (using correction factors)
12.	Flow Absolute Accuracy	± 2.0% ACFM	no correction factors flow >40 l/min
13.	Weight	350 lbs.	
14.	Power	1500 watts	while pumping
15.	Source	Carbon-14	
16.	Source Activity	100 microcuries	

2.0 TECHNICAL DISCUSSION

2.1 Principles of Operation

2.1.1 Beta Gauging

The Vehicle Exhaust Particulate Emission Gauge utilizes a filter "cassette" on which to collect the particles. Detection of particles on the filter is accomplished by an isotopic radioactive source (Carbon-14) and a scintillation detector (Photomultiplier Tube).

The attenuation of beta particles through a medium follows an exponential law:

$$N = N_o e^{-\mu\rho x} \qquad (1)$$

where N = detected counts

N_o = detected counts in absence of medium

μ = attenuation coefficient of medium

ρx = basis weight of medium.

In this application, the "medium" discussed consists of several separate media: 1) the air gap between source and detector; 2) the clean filter; 3) the detector window, and 4) the particles collected. Therefore equation (1) can be rewritten as:

$$N = N_o e^{-(\mu\rho x)_a} e^{-(\mu\rho x)_f} e^{-(\mu\rho x)_w} e^{-(\mu\rho x)_p} \qquad (2)$$

where N = detected count

N_o = counts emitted from surface of source in direction of detector

$(\mu\rho x)_a$ = attenuation exponent for the air

$(\mu\rho x)_f$ = attenuation exponent for clean filter

$(\mu\rho x)_w$ = attenuation exponent for detector window

$(\mu\rho x)_p$ = attenuation exponent for particles.

Obviously, the only factor of interest in Equation (2) is the last one referring to the particles. Every effort has been made to either control or minimize the effect of these other variables. The effects of variations in the density of the air has been minimized by reducing the air gap between the source and detector to about one centimeter. In order to account for variations between filters and to allow determination of the reduction in total collected counts due to detector attenuation, a standardization measurement is made and retained for later comparison. Equation (2) may then be written as:

$$N_2 = N_1 e^{-(\mu\rho x)}$$

where N_2 = detected counts after collection of particles (Measurement)

N_1 = detected counts before collection of particles (Standardization)

Taking the natural logarithm of both sides:

$$\text{Ln}\left(\frac{N_2}{N_1}\right) = -\mu\rho x$$

or $$\text{Ln}\left(\frac{N_1}{N_2}\right) = \mu\rho x$$

then $$\rho x = \frac{1}{\mu} \text{Ln}\left(\frac{N_1}{N_2}\right) \qquad (3)$$

Equation (3) is the expression to determine the mass per unit area collected. To determine total mass collected, multiply by total filter sample collections area A.

$$\text{Mass} = A\rho x = \frac{A}{\mu} \text{ Ln } \left(\frac{N_1}{N_2}\right). \quad (4)$$

Notice that by making an initial standardization measurement, all reference to the filter itself has been removed. This is indeed one of the advantages of the beta gauge. Almost any filter medium may be used, the only requirement being that the medium is not modified during the sampling process. The "cassette" approach accentuates this advantage, allowing use of filters not suitable for use as a tape.

The total gas flow through the unit is determined by use of a Fischer and Porter Swirlmeter (1" size). This unit creates a precessing vortex in a region containing a thermistor. The thermistor responds to the different heating afforded by the center of the vortex and, through pulse shaping, produces a pulse output. One pulse is representative of a specific volume of gas. To determine the total flow, the gauge simply collects the pulses from the flowmeter and multiplies by a constant. A more complete description is presented in the Operation Manual accompanying the gauge. The volume then becomes:

$$\text{Volume} = K_1 N_3. \quad (5)$$

To determine concentration, Equation (4) is divided by Equation (5):

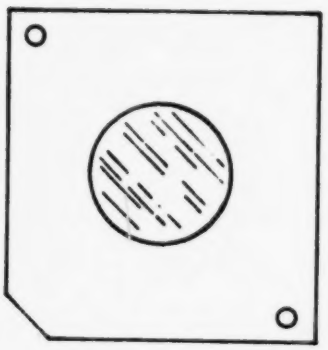
$$\text{Concentration} = \frac{\text{Mass}}{\text{Volume}} = K_2 \quad \frac{A}{\mu N_3} \quad \text{Ln} \left(\frac{N_1}{N_2} \right) \qquad (6)$$

where K_2 is a constant relating the various units involved. Equations (4), (5), and (6) represent the outputs required by contract. These along with the counts N_1, N_2, and N_3 are printed on paper to provide a permanent record of the test. By virtue of the Hewlett-Packard computer, all calculations, including the natural logarithm, are done digitally, affording extremely high accuracy of computation. The only analog circuitry in the gauge is the pulse gathering and shaping circuitry and the automatic gain control.

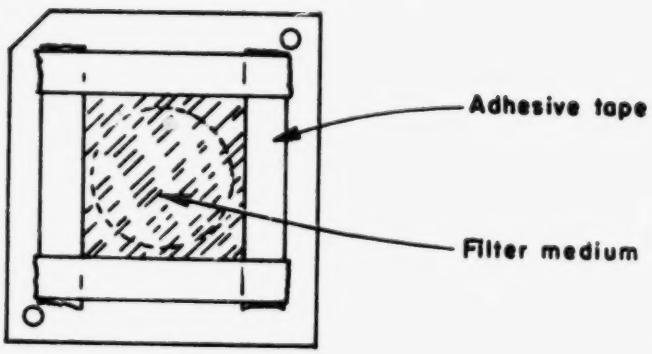
2.1.2 Cassette Transfer Mechanism

It has been previously mentioned that this gauge uses a filter cassette on which to collect the particles (See Figure 2-1). The cassette is constructed of 1/16" aluminum and is irregularly shaped to avoid improper loading into the magazine. Installation of the filter on the cassette is accomplished by cutting a square piece of filter paper (nominally 1" square) and taping the filter to the cassette. Notice also that the cassette has two holes on opposite corners to precisely locate the cassette at the beta measurement station. This is to insure that the source is irradiating the same area after collection on which standardization measurement was made before collection.

During operation each cassette is transported automatically through the gauge. The cassette transfer mechanism has three stations (Figure 2-2). The first is the loading station, where the lowest cassette



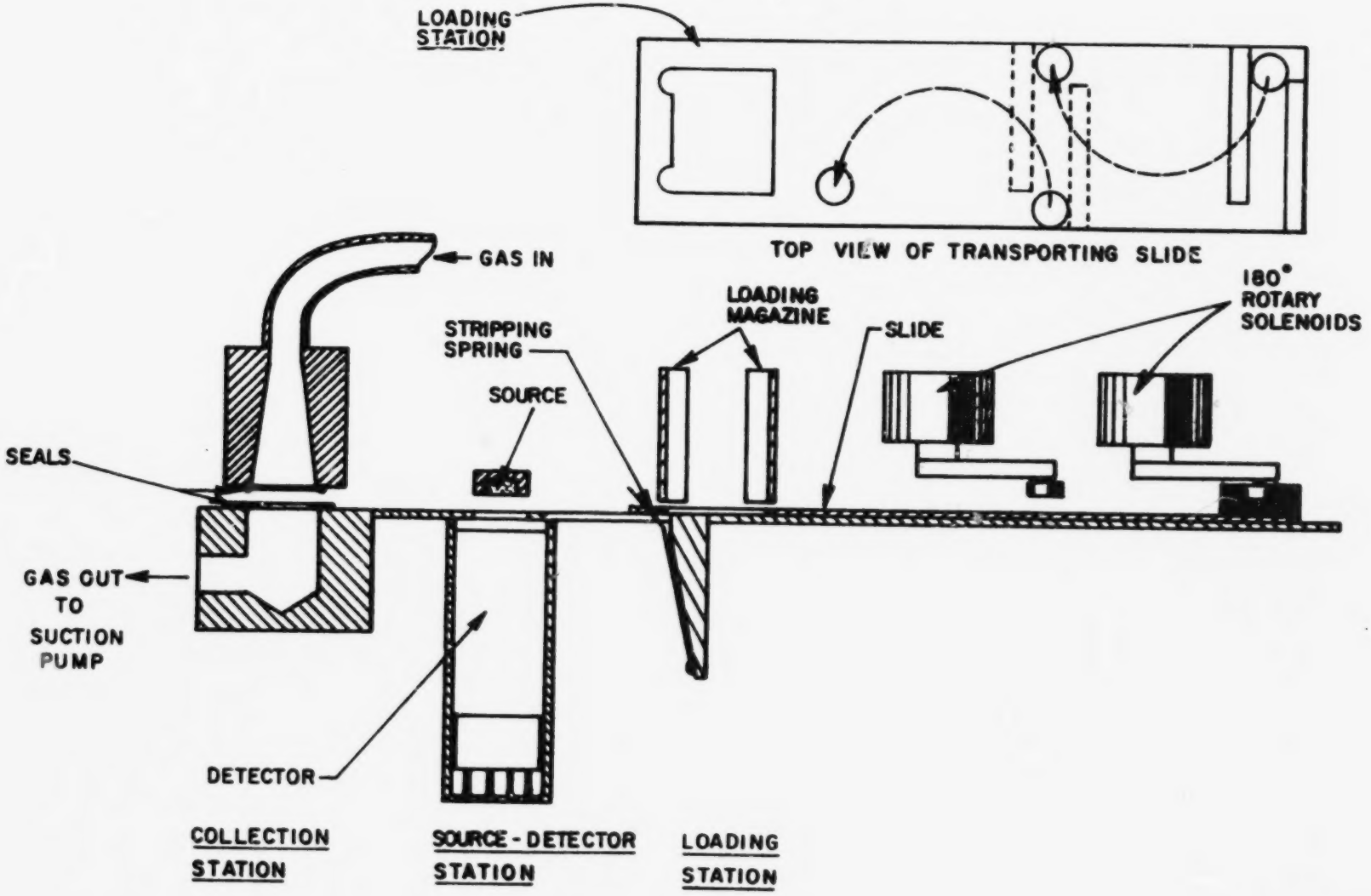
UPPER SIDE



UNDER SIDE

FILTER CASSETTE

Figure 2-1



SCHEMATIC LAYOUT OF THE THREE STATIONS & THE TRANSPORTING MECHANISM

Figure 2-2

in the magazine drops into a pocket in the transporting slide. At the second station the cassette is positioned between the source and the detector, so that the beam of radiation passes through the filter. At the third station the cassette is positioned so that the gas to be monitored may be drawn through the filter, and the particles collected.

Each cassette is transported between the stations by a slide of the same thickness as the cassette. The slide is moved between stations by two 180-degree rotary solenoids. In Figure 2-2 the slide is shown in its rightmost position, ready to pick up a cassette from the magazine. The next operation is for the right-hand solenoid to rotate 180 degrees. The cam roller on the end of the solenoid arm will follow the path indicated, causing the slide to travel to the left and transport the cassette to the source-detector station. At this station two tapered pins are pushed down into the two small holes in the cassette. At this time the standardizing measurement is made to determine a background count in the absence of particulate matter. The locating pins are then withdrawn and the left-hand solenoid is rotated 180 degrees. The cam roller on the end of the solenoid arm will follow the path indicated, causing the slide to travel further to the left and transport the cassette to the collection station. At this station the cassette is clamped between two elastomeric seals which prevent leakage of gas into or out of the stream passing through the filter. The cassette is then clamped, the vacuum pump is turned on, and the particles

are collected on the filter. At the completion of the collection cycle, the sealing clamp is released and the left-hand solenoid is rotated 180 degrees in the opposite direction to transport the cassette back to the source-detector station. Here the locating pins are reintroduced into the small holes in the cassette.

After the measurement of the material deposited on the filter is made, the right-hand solenoid is rotated 180 degrees in the opposite direction, and the slide is returned to its initial position. As it does so the cassette in its rightward travel encounters the stripper springs, two inclined flat springs which guide the cassette downwards through an opening in the base plate and out of the transporting slide pocket. The used cassette drops into a receiving container and the slide pocket is left vacant, ready to pick up another cassette from the magazine.

Originally, it was proposed that the transfer mechanism be a wheel in which holes had been cut to contain the cassette. After the program started, a more complete examination of this approach revealed certain problems. The problems, though not insurmountable, presented complex switching and sensing conditiongs which were considered to be simplified by using a linearly translating slide. If the filter manufacturing quality control were such that the basis weight from filter to filter did not vary by more than 10 $\mu g/cm^2$, a considerable reduction in complexity could have been accomplished. Current state-of-the-art in filter manufacturing does not permit this, however.

2.2 Design Criteria for Selecting the Filter Medium

Several filter media were evaluated during the course of the program. Before discussing the various filters, however, it is worthwhile to discuss the criteria for evaluation.

The contract required determination of the particulate concentration in a stream having at least 125 micrograms per cubic meter. This determination was to an accuracy of $\pm 25\ \mu g/m^3$ at a 95% confidence level and an efficiency of collection 99.9%. These requirements place a rather severe constraint on the sampling head repeatibility and the electronic system stability. In determining the amount of material which must be collected to allow this determination, consideration must be given to the magnitude of such error sources as the change in air gap conditions and filter deformation.

2.2.1 The Air Gap Effects

The air gap between source and detector represents a "background" attenuation for the beta particles. It is an attenuation which cannot be avoided and any variation in the air gap to cause a change in air density between standardization and measurement represents an error in measurement. To minimize the effect of air density variations, the air gap itself must be reduced as far as it practically feasible. In this gauge the air gap is about one centimeter. This represents an effective background basis weight of about $1.3\ mg/cm^2$. If the variation in air density can be held to within 1% (about $5^{\circ}F$) between standardization and measurement, then

one may consider ± 0.5% or ± 7 $\mu g/cm^2$ fluctuation (1σ) about a mean value of 1.3 mg/cm^2. Needless to say, if the variations are held to a closer tolerance, this error is reduced accordingly.

2.2.2 Filter Variation Effects

A filter mass normalization measurement is made prior to collection of the sample to eliminate the potential error due to non-uniformity of the filter material. However, since the filter is subjected to a reasonably high differential pressure during the sampling, it may deform somewhat and thereby change its mass per unit area. A mesh backing minimizes this effect. However, if an effective filter mass change of 0.1% of an original value of 3 mg/cm^2 occurs, a 1σ error of ± 0.1% or ± 3 $\mu g/cm^2$ may occur.

Another problem which must be considered is the property of almost all fibrous material to absorb and lose moisture during the process. It is unclear whether the property of retaining moisture is absorption, adsorption, or entrainment as these three phenomena often interrelate. Therefore in the remainder of this report, this property will be calll absorption. IN has had considerable experience with the problem of moisture in the manufacture of paper, and can say with certainty that moisture absorption or evaporation, while difficult to predict, can often represent a significant portion of a paper's basis weight.

In Figure 2-3, for example, are the results of an experiment using Pallflex Corporation's medium E70/2075W, a multi-layer medium using glass fiber to achieve high collection efficiency and a cellulose backing

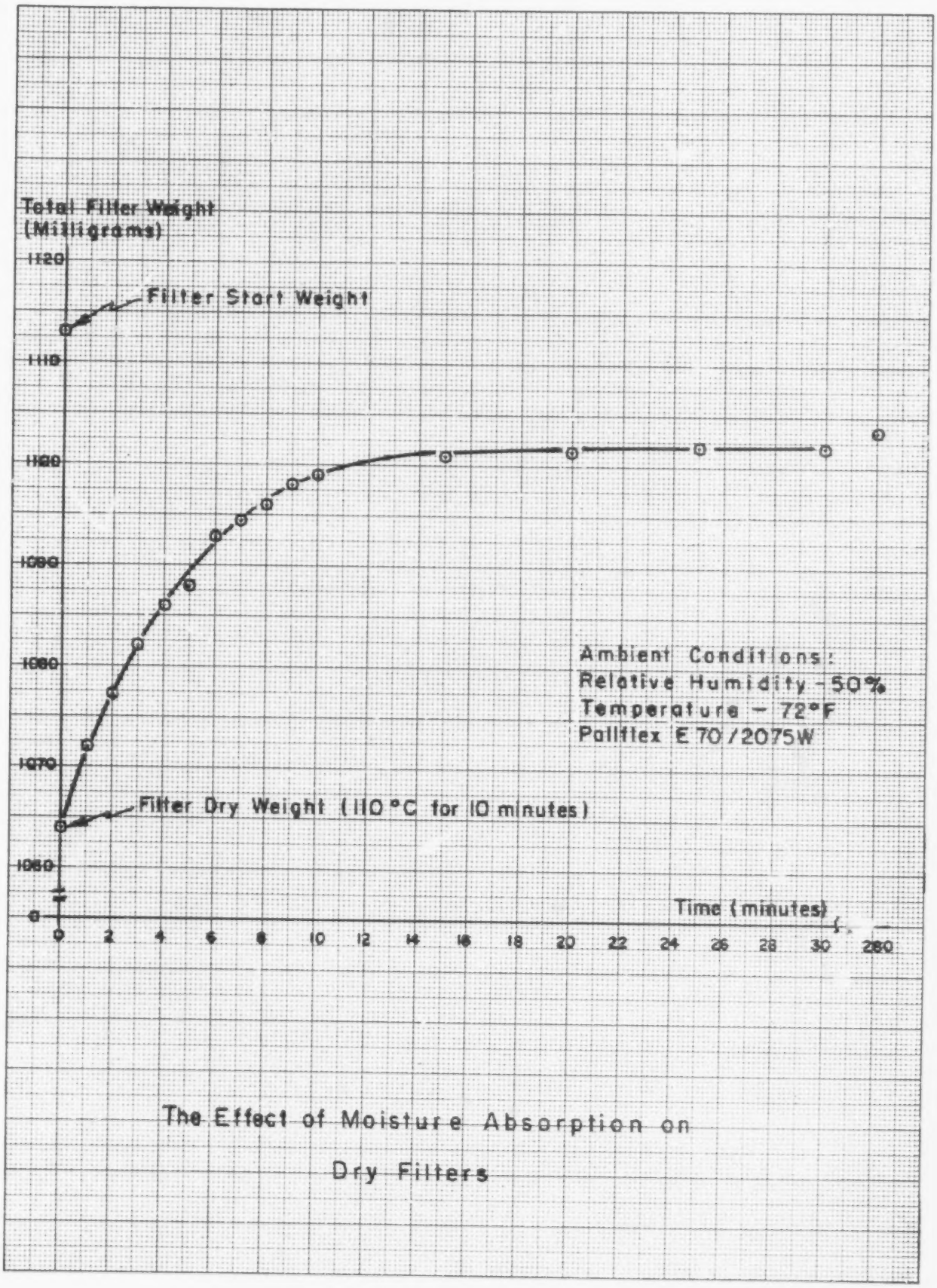
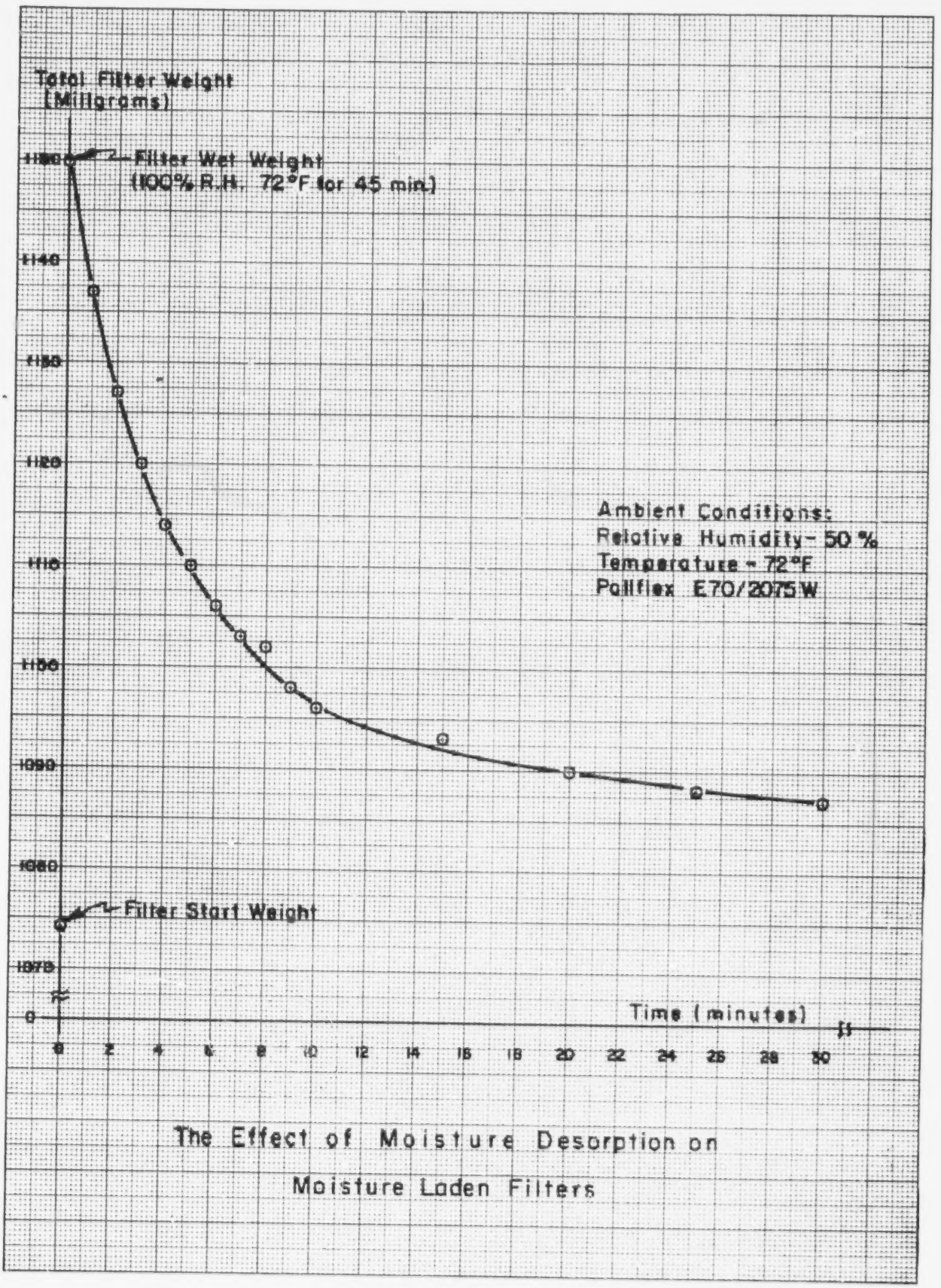

Total Filter Weight
(Milligrams)
1120
1110
1100
1090
1080
1070
1060
0
Filter Start Weight
Filter Dry Weight (110 °C for 10 minutes)
Ambient Conditions:
Relative Humidity - 50%
Temperature - 72°F
Pallflex E 70/2075W
Time (minutes)
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30 280
The Effect of Moisture Absorption on
Dry Filters

Figure 2-3

for extremely high structural strength. It has a basis weight of approximately 3.2 mg/cm^2. The sample chosen was a strip 31.8 cm x 110 cm. (This is equivalent in area to a circular filter approximately 210 mm in diameter. While admittedly large, an approximate number for any other size filter may be obtained by scaling the areas, as the variation on a basis weight consideration should be identical, regardless of filter size.)

The test procedure was to weigh the filter as delivered from the manufacturer, dry the filter at 110°C for 10 minutes (to obtain the actual "bone dry" weight would require much longer times such as 24 or 48 hours at 110°C), and then weigh the filter at periodic intervals after exposure to 72°F and 50% relative humidity. As can be seen, a weight loss of 49 milligrams (140 μg/cm^2) occurred upon drying. Much of this loss is recovered rapidly, so that after 10 minutes only 14 mg (40 μg/cm^2) remains unrecovered. At this point, however, the recovery becomes quite slow, so that after 30 minutes, 11 mg (31.5 μg/cm^2) still remain lost, and after 4 hours, 40 minutes, 9 mg (25.7 μg/cm^2) of moisture still had not been regained. For a typical filter size used (47 mm) for comparison, this would represent an error of 445 micrograms due solely to moisture absorption effects.

The effects of moisture desorption after having been exposed to 100% relative humidity at 72°F are even more dramatic (See Figure 2-4). After exposure of the filter to this environment for 25 minutes, a weight gain of 76 mg (217 μg/cm^2) as observed. After 10 minutes of



The Effect of Moisture Desorption on Moisture Laden Filters

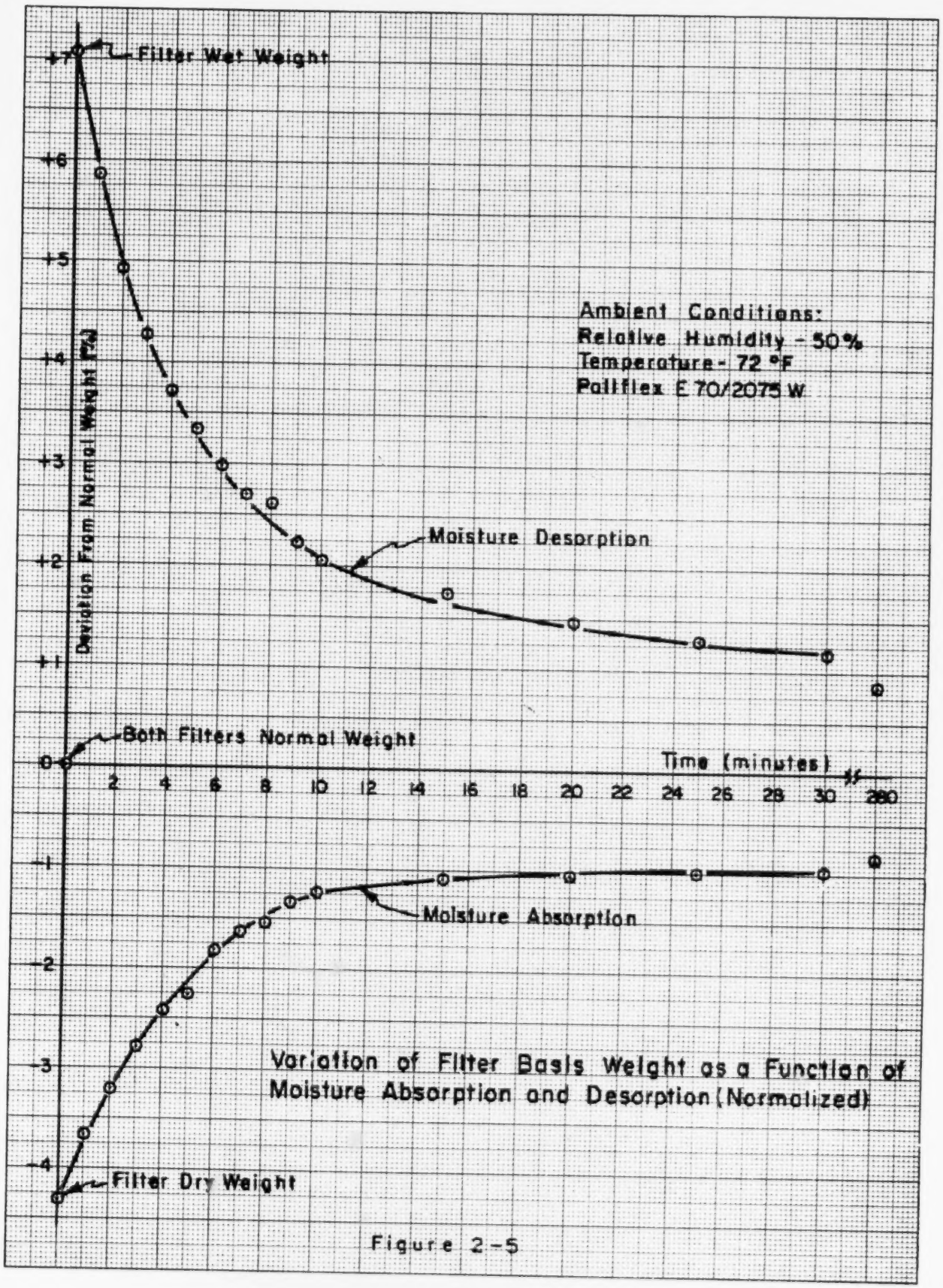
Figure 2-4

stabilization, 22 mg (63 $\mu g/cm^2$) were still retained, after 30 minutes, 12.8 mg (36.6 $\mu g/cm^2$), and after four hours, 40 minutes, there was still 9 mg (26.7 $\mu g/cm^2$) of excess moisture.

If the data is normalized to initial filter weight to present it on a percentage variation basis, Figure 2-5 is the result and illustrates clearly the "hysteresis" effect of moisture on fibrous materials. This hysteresis would represent an uncertainty in the amount of particulate mass collected of as much as ± 1% of the initial filter basis weight unless the complete moisture history is known. For the Pallflex medium, this is an error of approximately ± 32 $\mu g/cm^2$ due solely to moisture effects. In addition, if the measurement is made immediately after sampling (as it is in the case of this gauge), even higher errors can result.

From the above data, it is recommended that the Pallflex medium not be used in this gauge. There are, of course, other filter media which may be used which exhibit less sensitivity to either moisture absorption or desorption.

The GE Nuclepore, for example, showed a weight loss of only 6.4 $\mu g/cm^2$ after drying and recovered to its initial weight (no apparent hysteresis) after only three minutes of stabilization. When exposed to high humidity, it gained only 1.3 $\mu g/cm^2$ and has regained its initial weight within one minute. Unfortunately, this material does exhibit the highest pressure drop for a given flow rate of almost any other type of medium, and has shown a tendency to clog up.



Variation of Filter Basis Weight as a Function of Moisture Absorption and Desorption (Normalized)

Figure 2-5

The Gelman Type A filter avoids the problem of high pressure drop for a given flow rate and does not clog easily. Upon drying as previously described, this medium showed a loss of only 9.7 $\mu g/cm^2$ and has recovered half this amount in 15 minutes. When exposed to high humidity, however, this medium showed a weight gain of 221 $\mu g/cm^2$, but had regained its start weight within 10 minutes. It is also quite thick (approximately 7 mg/cm^2) and will require a longer counting time (see the next section).

A further consideration is that these errors were produced under static conditions (i.e., no forced flow through the filter) and may not be reproducible under dynamic conditions with which the EPA is involved.

2.2.3 Effect of Filter Thickness on Counting Time

The following discussion concerns only stream concentrations of 125 $\mu g/m^3$ as far as the flow rates are concerned. It is equally applicable when the required particulate basis weight resolution is of the same order.

In the process of a quantitive determination of the collected mass, the filter interferes with the measurements. For example, if the filter weighs 1,000 to 10,000 times the weight of the minimum desired resolution of the collected particulate, the total weight of the filter/particle system has to be determined to 0.1 to 0.01% accuracy. Even though it is feasible to determine the weight of certain objects to these accuracies, extreme precaution is required. The weight of an object like a paper or glass fibre filter is "unstable" in the sense that its weight will change with the

temperature and relative humidity of the environment and furthermore a hysteresis effect is observed in the adsorption and desorption of water vapor (see the previous section for a more complete discussion). Since the gain and loss of the weight with the environment and its history will be proportional to the weight of the filter, it is highly desirable to use as thin a filter as possible for a sample collection medium.

In a nucleonic method of weight determination, a further consideration is required in the sense that the filter will attenuate the radiation without contributing to the signal. This attenuation is not detrimental except for the possible physical deformation of the filter due to pumping, if no restriction is imposed on the required source strength. However, if the source strength is limited to a given value, the loss of radiation intensity has to be made up by a long counting time to obtain a desired statistical accuracy.

Consider a case of "zero" filter thickness. Then, with a 100 μCi C-14 source and the geometry of IN design, a count rate of 50,000 cps is obtained. If a two sigma resolution of ± 20 $\mu g/cm^2$ (corresponding to a stream resolution of ± 25 $\mu g/m^3$ with a 4 CFM sampling flow rate, sampling for 20 minutes from an aerosol stream containing 125 $\mu g/m^3$ of particulate) or ± 10 $\mu g/cm^3$ one sigma resolution is required, the required counting time is determined as follows: The attenuation of radiation is given by

$$N = N_o e^{-\mu\rho x} \qquad (7)$$

where N and N_o are the detected total counts with and without the sample; and μ, ρ, and x are the mass attenuation coefficient, density and thickness of the sample, respectively. By differentiation, one obtains

$$\frac{dN}{N} = -\mu\rho x \left[\frac{d(\mu\rho x)}{\mu\rho x}\right]$$

or

$$\frac{dN}{N} = -d(\mu\rho x). \tag{8}$$

For C-14 beta rays, μ is approximately 250 cm^2/gm and the minimum required resolution of (ρx) is ± 10 $\mu g/cm^2$ (one sigma). Therefore, the minimum required resolution for $(\mu\rho x)$ is $250 \times 10 \times 10^{-6} = 0.0025$. This means, from equation (8), that the nuclear signal, N, has to be resolved to an accuracy of $\pm 0.25\%$ (one sigma). It is well known that the nuclear signal fluctuates with one sigma value of $\pm \sqrt{N}$ when the total count is N. Therefore,

$$\frac{dN}{N} = \frac{\sqrt{N}}{N} = \frac{1}{\sqrt{N}} = 0.25 \times 10^{-2} \tag{9}$$

or

$$N = 1.6 \times 10^5. \tag{10}$$

Since the count rate with no sample, n_o, is 5×10^4 cps, the counting time to accumulate 1.6×10^5 counts is

$$T = \frac{1.6 \times 10^5}{n_o} = \frac{1.6 \times 10^5}{5 \times 10^4} = 3.2 \text{ seconds.} \quad (11)$$

Now, if the filter is assumed not to contribute further error due to its unstable weight (absorption and desorption of moisture) and physical deformation, its effect on the nucleonic measurement of collected particles is to attenuate the radiation. Therefore, the count rate, n_o, will be reduced according to

$$n = n_o e^{-(\mu\rho x)_F} \quad (12)$$

where the subscript, F, represent the value for the filter. Substituting n for n_o in equation (11), one obtains the required counting time for various filter thicknesses from

$$T = \frac{1.6 \times 10^5}{n} = 3.2\, e^{(\mu\rho x)_f} \text{ seconds.} \quad (13)$$

This equation is plotted in Figure 2-6 labeled with "no electronic error."

An additional error is caused in the measurement of N(for the purposes of discussion, this is called electronic error, but also includes all other errors besides statistics) in equation (7). Thus, 0.25% total allowed error in equation (9) should be allotted between the electronic error and the statistical fluctuation. Assuming an RMS averaging, one gets

$$E_e^2 + \frac{1}{N} = (2.5 \times 10^{-3})^2 \quad (14)$$

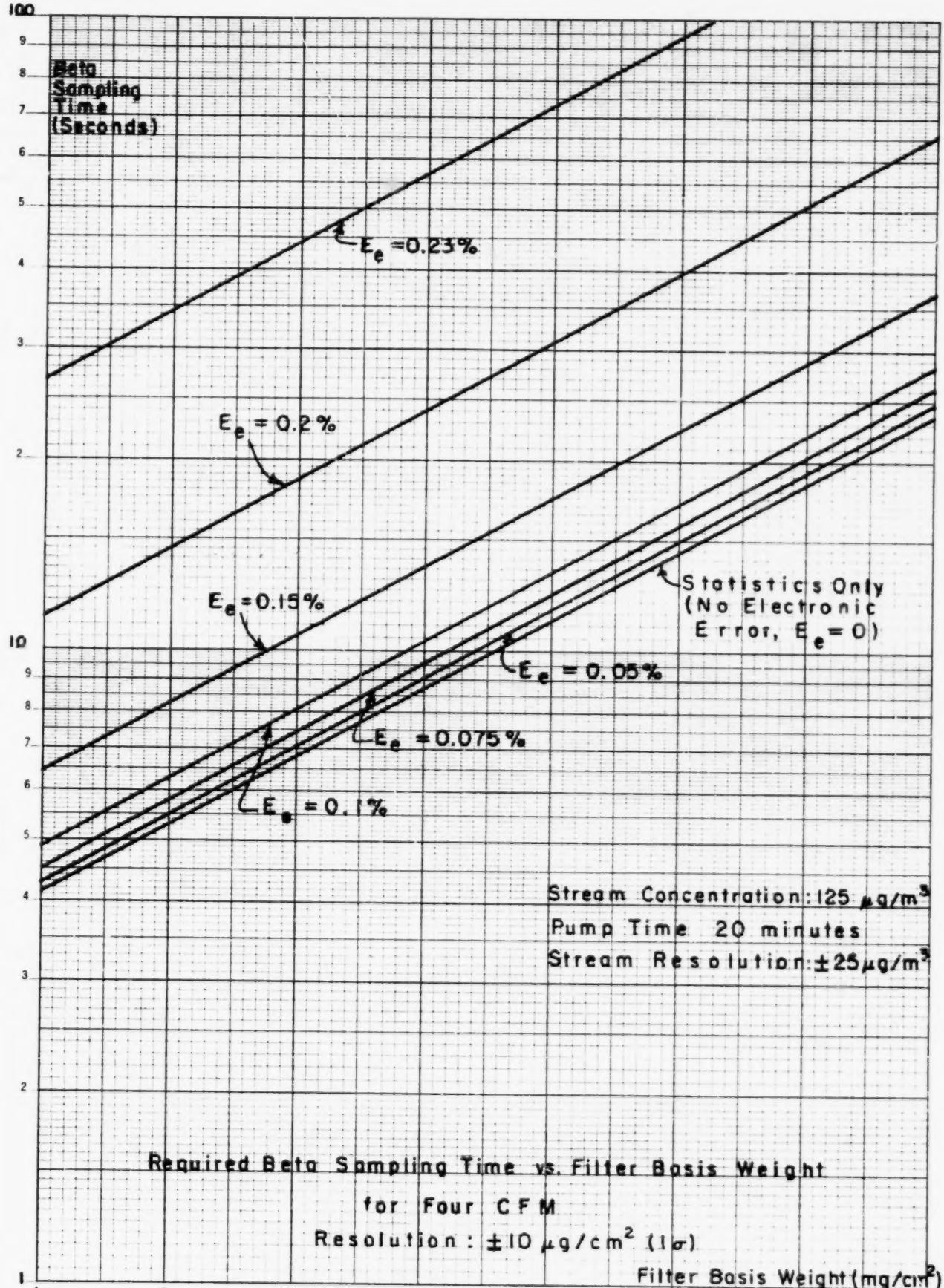

Beta Sampling Time (Seconds)
E_e = 0.23%
E_e = 0.2%
E_e = 0.15%
E_e = 0.1%
E_e = 0.075%
E_e = 0.05%
Statistics Only (No Electronic Error, E_e = 0)
Stream Concentration: 125 µg/m³
Pump Time 20 minutes
Stream Resolution: ±25 µg/m³
Required Beta Sampling Time vs. Filter Basis Weight for Four CFM
Resolution: ±10 µg/cm² (1σ)
Filter Basis Weight (mg/cm²)

Figure 2-6

or

$$N = \frac{1}{(2.5 \times 10^{-3})^2 - E_e^2} \quad . \tag{15}$$

Following the same procedure as in the case of "no electronic error," one obtains the counting time from

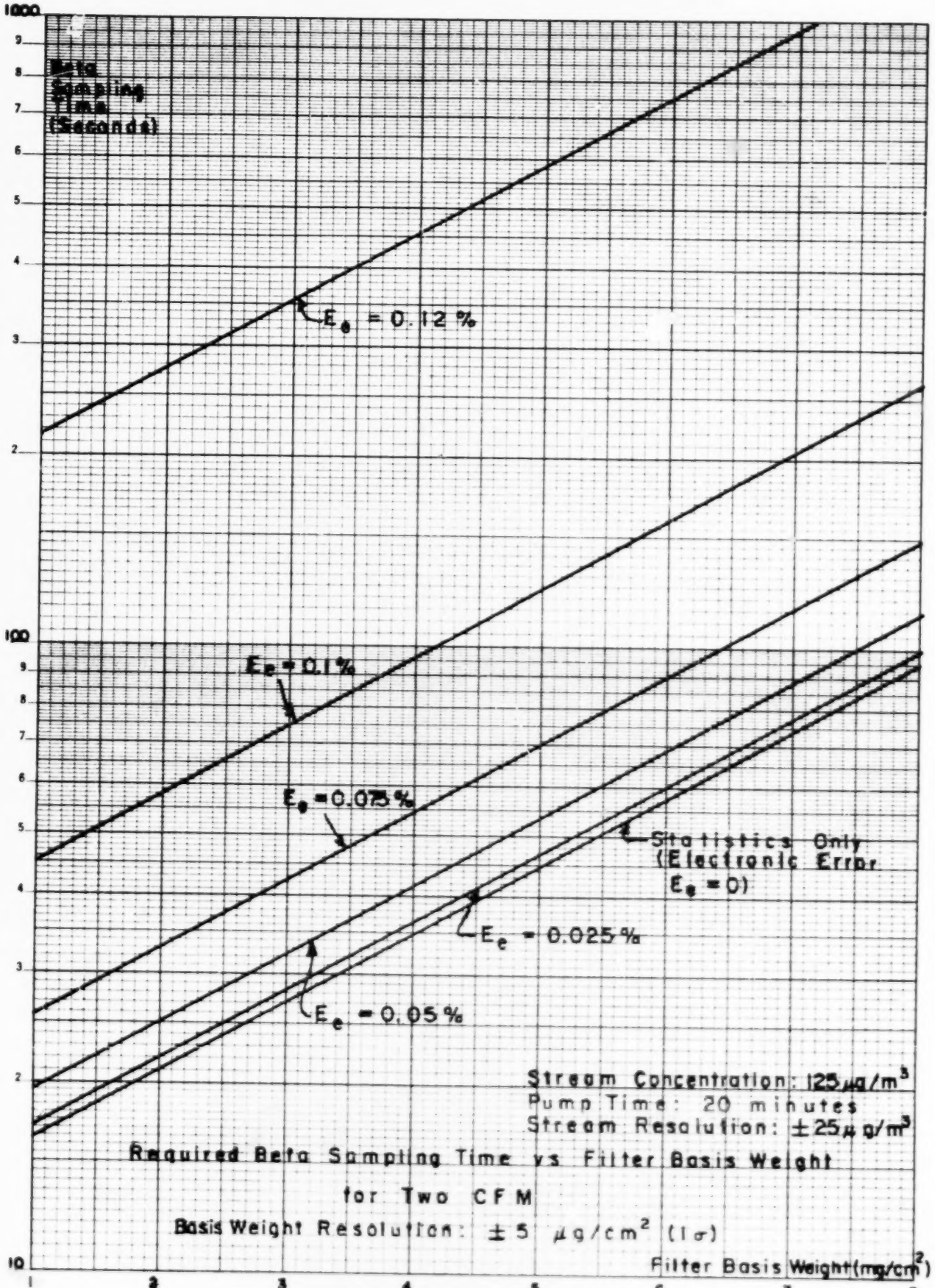
$$T = \frac{N}{n} = \frac{e^{(\mu\rho x)_f}}{5 \times 10^4 \left[(2.5 \times 10^{-3})^2 - E_e^2\right] \text{ seconds.}} \tag{16}$$

This equation is plotted in Figure 2-6 with E_e = 0.05%, 0.075%, 0.1%, 0.15%, 0.2%, and 0.23%.

If, however, the same stream resolution (± 25 $\mu g/m^3$) is required, but only 2 cfm (56 liters/min) flow rate is used, the basis weight resolution is ± 5 $\mu g/cm^2$ and Figure 2-7 is the result with E_e plotted for 0.025%, 0.05%, 0.1%, and 0.12%. As can be seen, the effect is rather dramatic on the counting times, with resolution of this magnitude impossible at 4 cfm if all other errors amount to 0.25%, and at 2 cfm if all other errors amount to 0.125%.

2.3 General Comments on Filter Selection

While the advance copy of the Operation Manual delivered with the instrument recommended that the Pallflex E70/2075W medium be used, later test results show this medium to be undesirable from a moisture absorption and desorption standpoint. It is IN's opinion that the "optimum" filter medium has not yet been found. Much work remains to be done in this area. In selecting a medium to use, however, several conclusions can be drawn from the preceding sections of this report.



K+E SEMI-LOGARITHMIC 46 4973
2 CYCLES X 70 DIVISIONS MADE IN U.S.A.
KEUFFEL & ESSER CO.

Figure 2-7

First, the filter should have as low a basis weight as possible for two reasons. One, to reduce the amount of time required for beta measurement, and two, to reduce the effect of any slight distortion caused in the filter by the pumping process. From the previous section, the beta collection time may vary as much as four to one, depending on the basis weight of the filter. Also a 1% basis weight variation for 1 mg/cm^2 filters is 10 $\mu g/cm^2$, but for 8 mg/cm^2, a 1% basis weight variation is 80 $\mu g/cm^2$.

Second, the property of paper-based filters to absorb and lose moisture during the process casts doubt on results obtained using these materials. In fact, even the Gelman Type A filters yield dubious results after exposure to high humidity, which unfortunately happens to be the situation with automotive exhaust. This does, of course, mean that there is currently no standard against which to reliably measure results, either with the beta gauge or with gravimetric comparisons.

Third, the problem of moisture is not confined to beta gauging. In fact, it isn't even confined to moisture. Any volatile material would be subject to identical fluctuations. Again, automotive exhaust contains a high percentage of volatile material, at least in the cases of low-leaded and non-leaded fuels.

Finally, it is conceivable that a different type of filter material may be required from filter manufacturers. Little consideration has

been given by this industry to such properties as low moisture sensitivity. This is presumably due to primary application areas being chemical analysis of collected particulate or simply to clean the air passing through the filter.

2.4 Selection of Carbon-14/PMT Geometry

Several sources were evaluated prior to contract: Kr-85, C-14, Ni-63, and Pm-147. A complete discussion of the tradeoffs involved in selection is beyond the scope of this report and only the major reasons will be presented here. Krypton-85 was eliminated on the basis of poor sensitivity. Nickel-63 was eliminated on the basis of low modal energy. Promethium-147 was eliminated on the basis of short half-life. This left Carbon-14 as the optimum selection.

IN had proposed that the source to use was in the form of a carbide, where there is a chemical bond between the carbon and surrounding atoms. In so doing, an extremely high integrity source could be achieved, capable of surviving almost any environmental condition, up to and including fire. IN still maintains that this is the most desirable source to have. Unfortunately a complete survey of the source manufacturing field disclosed no vendor willing to undertake the development of a carbide source. While the technology appears to be available to produce such a source, neither time nor money would permit the development under this contract.

The vendor eventually selected was New England Nuclear, Billerica, Massachusetts. Originally a source was received from them which was a C-14/Kapton mixture deposited on a small aluminum disc. This proved to be unsatisfactory from a uniformity standpoint. Several iterations were required before achieving the physical integrity, uniformity, and emission efficiency we desired. IN believes that the source which is now in the gauge is one of the finest non-carbide sources made. It is constructed by depositing the C-14 on an aluminum planchet, laying a thin plastic film over this and spraying the active area with Kapton. This source has the highest percentage yield of any other in IN's experience and made it possible to reduce the activity to such a level as to prove the feasibility of making this gauge eventually license free.

2.4.1 Determination of Source Strength Requirement

The license free aspect of the gauge was paramount in IN's philosophy of design. Carbon-14 occurs naturally and as a result has one of the highest allowed quantities to qualify for National AEC license exempt status. This quantity is 100 microcuries. It has already been discussed (Section 2.2.3) how much collection time is involved, providing a count rate of 5×10^4 pulses per second can be achieved in the absence of a filter. It will now be shown that this count rate is in fact achieved in this gauge.

The effective yield of the 100 microcurie source delivered with the gauge is estimated by New England Nuclear at 15%. The number of counts actually emitted from the source in all directions is

$$3.7 \times 10^{10} \ \frac{\text{counts}}{\text{sec.} \cdot \text{Curie}} \times 10^{-4} \ \text{Curies} \times .15 = 5.5 \times 10^{5} \ \frac{\text{counts}}{\text{second}} .$$

If the source is place 1 cm from a disc-shaped detector of 1 inch diameter, the geometrical factor is $1/2 \ (1 - \cos \Theta) \simeq 18\%$ where Θ is the half-angle subtended by the detector. Thus, in the absence of air and sample $5.55 \times 10^{5} \times .18 = 10^{5}$ counts per second reach the detector.

The effective attenuation coefficient (μ) has been estimated at 250 cm^2/gm. The desired count rate is in the absence of the filter but the attenuation due to air and the detector window must still be considered. It was previously discussed that beta radiation attenuation obeys a negative exponential law:

$$N = N_o e^{-\mu \rho x} .$$

This can be expressed differently as

$$N = N_o e^{-(\mu \rho x)_A} \ e^{-(\mu \rho x)_W}$$

where

$N_o = 10^{5}$ counts per second (previously derived)

$(\mu \rho x)_A$ = attenuation exponent due to air

$(\mu \rho x)_W$ = attenuation exponent due to the detector window.

For air, the density is equal to approximately 1.2 mg/cm^3 and the distance is 1 cm. The attenuation factor ($e^{-\mu\rho x}$)$_A$) due to air then becomes 0.74. The detector window has a basis weight (ρx) of approximately 0.9 mg/cm^2 (manufacturer's specification). Therefore the detector window attenuation factor is 0.8. The total detected counts then becomes $10^5 \times .74 \times .8 = 5.92 \times 10^4$ counts per second. Allowing for detector efficiency and inclusion of a threshold detector to exclude PMT characteristic noise, it can be seen that the detected counts will not vary significantly from the 5×10^4 counts per second required. In fact, the count rate achieved in the absence of a filter is approximately 44,000 cps so that good agreement with theory is achieved.

This count rate allows the use of the gauge over a wide range of filter area densities without inordinately increasing the beta measurement time (See Section 2.2.3). This was deemed desirable by IN in order to increase its flexibility as a research tool. In Figure 2-6, for example, it can be seen that using a filter of basis weight 7 mg/cm^2 (such as the Gelman Type A) and a process repeatibility of 0.1% (estimated reliability for the IN gauge) a counting time of 220 seconds is required to achieve ± 10 μg/cm^2 resolution. If the count rate were one-tenth our estimated rate (this would yield an estimated detected count rate of 4.4×10^3 counts per second, typical of maximum rates available from Geiger-Mueller detectors), this time would be 2.200 seconds to achieve the same accuracy.

2.3.2 Detector Selection

The deficiencies of a Geiger-Mueller tube in the rapid determination of filter basis weight has been discussed. There are three other types of detectors which may be used for low energy beta radiation detection: solid state detectors, proportional counters, and a PMT/scintillation crystal combination. The current state-of-the-art in solid state detectors are such that they must be used in a nearly constant temperature environment. This was felt to be an unnecessary requirement to place on the detector station. Proportional counters and ionization chambers were considered carefully, but the window thickness is such as to prevent the use of license exempt quantities for C-14 sources. That left only the PMT/scintillation crystal combination.

There are two basic scintillation crystals which could be used: CaF_2(Eu) or NaI(Tl). Since the modal energy of C-14 is so low, an extremely thin window is required (on the order of 1.0 mg/cm^2). For this reason and the fact that NaI is highly hygroscopic and would tend to deteriorate with time, CaF_2 was selected as the scintillator. Harshaw Chemical Company was then contracted to fabricate the detector unit, using an Amperex XP-1011 tube, a CaF_2 crystal, and aluminized Mylar as the detector window.

This detector was quite stable in gain in contrast to other very similar detectors. As is the case with all PMT's, however, the gain drift was significant enough to require the addition of an automatic

gain control (AGC) circuit. A typical beta spectrum is as shown in Figure 2-8. If two thresholds are established, L and H for low and high, respectively, it is possible to select these thresholds such that the difference in the two numbers remains relatively constant. For any major changes which may occur, however, the ratio of the two counts provides a control signal by which the gain can be adjusted to keep the above condition true. Through this stabilization technique, PMT stabilities on the order of 0.1% are achieved and it is this capability that allows the measurement to be made at the low end of sensitivity.

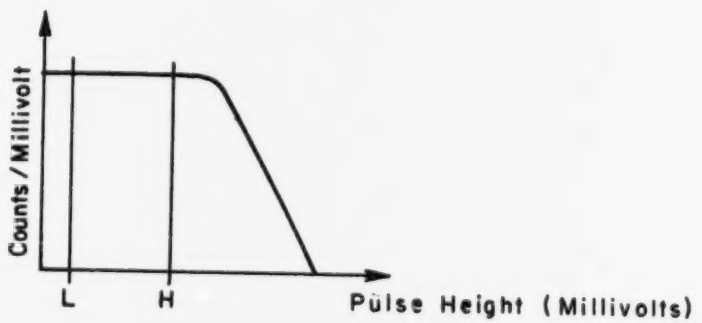


Figure 2-8

3.0 FIELD TESTS

Two series of tests were performed on the gauge. The first series was performed during the week of 4 October 1971, at the EPA's Cincinnati facility and was for the primary purpose of debugging the unit. The second series was performed during the week of 25 October 1971, at the Dow Chemical Company in Midland, Michigan, and was for the primary purpose of acceptance tests on the unit. The results obtained from both these series are the subject of this section of the report. Prior to the field tests, the attenuation coefficient μ was experimentally determined to be 252 cm^2/gm by noting the effect on the count rate of differring thickness of Mylar.

3.1 The Cincinnati Series

Little data was obtained from automotive tests in this series due to several problems encountered with the unit. As the purpose of the tests was to reveal these problems, however, considerable progress may be considered to have been made. Minor problems were encountered with the locating solenoid binding up. Due to self-check microswitches installed on the unit, no errors in data due to this malfunction are believed to have occurred. A problem was encountered but not recognized until after the Dow series with interference between the pressure fluctuations in the air line due to the pump and the Swirlmeter. Apparently, these fluctuations were viewed by the Swirlmeter as vortex precessions and counted as a volume of flow. It is believed that this was only a problem

at the low flow rates. To correct the problem, the flowmeter was changed from its "downstream of the pump" location to its present "upstream of the filter" location and the interference was no longer observed. During the brief redesign period between the Cincinnati and Dow series of tests, all encountered deficiencies were corrected.

One of the major thrusts of this series was to evaluate different filter materials. There were four different media which were hopefully to be tested: 1) the Gelman Type A; 2) GE Nuclepore (0.4 μ and 1.0 μ), 3) Pallflex No. E70/2075W; and 4) a paper filter of high efficiency and low basis weight developed by Dr. Wendell Anderson, Naval Research Lab., Washington, D. C. Details of the tests follow, but a brief discussion of these media is appropriate. Some deformation (tearing) of Dr. Anderson's material was detected. This is believed to be a result of the short time delay between the command to release the clamp on the cassette at the sampling station and the command to transport the cassette to the beta measurement station. After this, it was decided to switch to the Pallflex medium, and as this filter seemed to perform adequately at the time, no tests were performed on either the Gelman or GE filters. Tests were later performed which showed the Pallflex medium to be inadequate due to moisture interferences (See Section 2.2.2) and it is now recommended that tests be performed by the EPA on the Gelman and GE filters to determine their adequacy.

This test series can be divided into six separate segments. The included segments, reasons for their inclusion, and results obtained are contained in the following sections. In this entire series, volume collected is unimportant and will not be included in the data.

3.1.1 Clean Air Tests

These tests were performed on Dr. Anderson's medium and were for the purpose of determining the clean air characteristics of the unit. Clean air was obtained by inserting an in-line 47 mm Gelman Type A filter in the air line at the entrance port to the unit. Only two tests were performed. In both, the beta collection time for standardization and measurement was 90 seconds. In test 1, the pumping time was 23 minutes; in test 2, the pumping time was 5 minutes.

Test 1 indicated a total collected mass of 432.8μg while test 2 indicated a total collected mass of 169.5μg. Both of these values were unacceptable since the gauge specification was ± 30μg for 2 cfm flow rate and ± 60μg for 4 cfm flow rate and this latter flow rate was the maximum attainable with the unit. It was suggested that the cassettes may introduce errors and the next group of tests were devoted to seeing if that error was constant.

3.1.2 Cassette Effects

Seven cassettes were labeled A through G and installed in the unit with no filter attached. Here are the results:

Cassette No.	Total Indicated Mass (Micrograms)
A	+ 3.9
B	+ 393.1
C	+ 358.3
D	- 42.9
E	+ 243.6
F	+ 249.6
G	+ 82.7

These tests showed that the error, if cassette introduced, was at least not constant for all cassettes. It was then suggested that possibly the temporary interruption of the beam by the cassette as it moved into position may cause a disturbance of the AGC. To determine this, a delay was introduced between the time the cassette was moved into the source-detector station and the time the computer began accepting data.

3.1.3 AGC Delay Effects

Four of the lettered cassettes used in the previous test were again tested with these various delays. Here are the results:

Cassette No.	Delay (seconds)	Total Indicated Mass (micrograms)
B	60	- 191.3
C	15	- 44.4
D	15	- 88.9
G	60	+ 125.5

While the grouping is tighter and generally of opposite sign from the previous tests, these data were judged inconclusive and unacceptable. It was then decided to investigate the performance of the gauge without either filter or cassette in the transfer mechanism.

3.1.4 Stability Tests

These tests were performed by placing the unit in "automatic" and allowing it to run with no cassettes in the loading magazine. During test 9, it was noted that the count rate being observed on the Nixie display of the Computing Counter exhibited a sensitivity to whether or not sunlight was shining on the floor of the test area. On tests 10, 11, and 12, precautions were taken to ensure that the external light reaching the gauge was minimized. On tests 13, 14, and 15, the front door of the unit was opened to allow light to shine on the cassette transfer mechanism. On test 16, the light was again shut off. Here are the result:

Run No.	Total Indicated Mass (micrograms)
1	-1343.6
2	- 50.6
3	+ 143.4
4	- 33.7
5	+ 100.8
6	- 26.6
7	- 183.1
8	+ 270.7
9	+ 131.4
10	- 59.6
11	+ 68.1
12	+ 49.6
13	+ 131.6
14	+ 470.0
15	- 301.4
16	- 21.1

These tests confirmed conclusively that a light leak did exist in the detector. In tests 10, 11, 12 and 16 the values obtained, while larger than desirable, were considered adequate for this series provided flow rates of approximately 4 cfm are used. It was then decided to use filters in the transfer mechanism, taking care to see that external light reaching the interior of the unit was minimized.

3.1.5 Stability Tests -- Using Filters

Tests were performed again using Dr. Anderson's medium on the clean air previously discussed. Up until this point little attention had been paid to deformation of the filter. On tests 5 and 6, filter deformation in the form of tearing of the filter away from the cassette was definitely observed.

Run No.	Total Indicated Mass (micrograms)
1	- 86.4
2	+ 48.1
3	- 3.0
4	+ 29.0
5	+292.6
6	+ 95.9

On the basis of these data, the decision was made to change media and use the tougher Pallflex media for the remainder of the tests. It was decided to repeat the previous test using the Pallflex medium.

3.1.6 Stability Tests -- Pallflex Medium

Tests were again performed using a 47mm in-line filter to obtain clean air. It had also been theorized at this point that some error may be occurring due to either moisture loss in the medium

or actual material loss from the filter itself. This was as a result of data from tests at IN prior to the Cincinnati test series that showed the no-cassette stability to be well within the ± 30μg total indicated mass specification. Thus six tests were run using the same filter cassette on all tests.

Run No.	Total Indicated Mass (micrograms)
1	-126.4
2	loss of record
3	- 83.2
4	- 58.6
5	- 31.4
6	- 27.4

On the basis of these data, it became fairly clear that some kind of process error was occurring. It was not determined at this time whether the error was due to moisture or material loss. Later studies (Section 2.2.2 of this report) point the finger rather strongly towards moisture, but no conclusion can be drawn from these studies regarding the data from either the Cincinnati or Dow series of tests.

This concluded the Cincinnati series except for some tests on a laboratory aerosol which were unsuccessful due to the shortness of time available. Even though limited data had been obtained, the series was considered successful in that the obvious deficiencies in design and alignment had been pinpointed.

3.2 The Dow Series

The tests at the Dow Chemical Company in Midland, Michigan, during the week of 25-29 October, 1971, may be divided into three

general areas: tests on 1) leaded fuels (3.0cc/gal TEL), 2) unleaded fuels (0.0 cc/gal TEL), and 3) low-leaded fuels (0.5 cc/gal TEL). Three different cars were used for the three different areas and will be discussed later. Tests were performed with the automobiles operating under steady state conditions (constant speed), the Federal LA-4 cycle, and the older California Seven-Mode cycle.

The Dow facilities were quite well equipped and similar in basic construction to the EPA's Cincinnati facilities. An isokinetic mixing tube was used, in which the overall flow was 500 cubic feet per minute. Two samples were taken by Dow for gravimetric comparison to the beta gauge. Initially these samples were two 143 mm filters but later consisted of one 143 mm filter and one 2 inch filter.

Prior to the actual automotive tests, a test was run using a cassette prepared with a filter as the medium (all tests but one used Pallflex E70/2075W) was delivered from the manufacturer. This filter showed a weight loss of 70.6 micrograms. In order to test the stability of the unit (± 30 micrograms was the specification) two tests were run with no cassette at all in the instrument showing +10.9μg and +4.1μg, respectively. Convinced that electronically the unit was well within specification, several cassettes were prepared by baking at various temperatures for various times and allowed to stabilize for various short times in the ambient conditions. This baking discolored the filter somewhat, possibly introducing contaminants (no discoloration had

ever been noticed before at idential temperatures), and as a result this conditioning was no longer employed after this series. The pump was then allowed to draw clean air through the filter for a short time. The results are as follows:

Test No.	Bake Time (min)	Bake Temp (^{o}F)	Equalize Time (min)	Pump Time (min)	Weight Change (micrograms)
1	10	210	10	5	-54.0
2	10	210	20	5	-93.2
3	10	210	30	5	-83.2
4	10	210	45	5	-55.4
5	20	200	30	20	-48.2
6	--	---	--	20	-77.8

Test No. 6 is a repeat of the cassette used in test No. 5 with no intermediate baking. That is to say, the cassette was immediately reprocessed through the beta gauge to determine whether the same phenomena which existed during the Cincinnati series (Section 3.1.6 of this report) could be duplicated here. Obviously, it was not.

The average of the above six tests was -68.6 micrograms, and represented a "process" error (since all readings are negative and in the 50 to 90 microgram category). Since no solution to this problem was immediately apparent, it was decided to delay a more detailed study in this area since the first automotive series would be run with leaded fuels and the mass of collected particles was expected to be much higher than this figure.

3.2.1 Tests on Leaded Fuel (3.0 cc/gal TEL)

On tests 1 through 8, the flowmeter was placed downstream of the pump. After these 8 tests, it was determined that the pressure fluctuations introduced into the air flow by the pump was interferring with the vortex precession action of the Swirlmeter. For all subsequent tests at Dow, the flowmeter was placed upstream of the filter.

A synopsis of this series is presented in Table 3-1. Little conclusion can be drawn from tests 1 through 8 due to pump interference. In tests 5, 9, and 10, it is suspected that there was electrical noise introduced into the power line, causing the computer to malfunction. This problem did not occur for the remainder of the Dow series. Tests 11 through 15, however, showed excellent agreement between the beta gauge and a gravimetric comparison.

3.2.2 Tests on Non-Leaded Fuel

The next series of tests were performed using an automobile which had been fueled solely with non-leaded fuel since its manufacture. Considerable discrepancy existed in cases between the beta gauge and the gravimetric comparison. This phenomenon is not clearly understood as of the date of this report. To provide additional data, and since good agreement was obtained between the two 143 mm gravimetric comparisons, it was decided to substitute a 2 inch diameter filter for one of the 143 mm diameter filters for test 20 and all subsequent tests.

TABLE 3-1

LEADED FUEL TESTS

Test No.	Test Mode	Pump Time (minutes)	IN Mass Gain (μg)	IN Total Flow (liters)	Dow 143 mm Gain (μg)	IN Rate (mg/min)	Dow Rate (mg/min)	% Dev. IN - Dow / Dow	Remarks
1	Moist. Chk.	5	-14	595	--	--	--	--	
2	30 mph	20	2980.	2315	3650	18.2	22.8	-20.1	A
3	30 mph	20	2491	1935	3850	18.2	24.1	-24.2	B
4	30 mph	20	2485	1840	3600	19.1	22.5	-15.1	B
5	--	--	--	--	--	--	--	--	C
6	60 mph	20	2366	874	13200	38.3	82.5	-53.6	
7	LA-4(C.S.)	23	3230	1022	14000	45.8	76.1	-39.8	B
8	LA-4(H.S.)	10	2151	787	4050	39.1	50.6	-22.7	
9	LA-4(H.S.)	23	--	--	--	--	--	--	C
10	LA-4(H.S.)	23	--	--	--	--	--	--	C
11	60 mph	10	3627	453	9150	113.3	114.4	- 1.0	D
12	60 mph	10	3870	795	5800	68.9	72.5	- 5.0	A
13	60 mph	10	1929	417	5750	65.5	71.9	- 8.9	
14	30 mph	15	1502	1184	2400	18.0	20.0	-10.0	
15	15 mph	15	1437	1382	1750	14.7	14.8	- 0.6	

NOTES:
1. Dow Flow Rate for all tests - 4 CFM
2. Mass Flow Rates are from automobile
3. Fuel is 3.0 cc/gal TEL
4. Vehicle is 1971 Chevrolet - 350 CID Engine

REMARKS:
A. Flow not constant throughout test
B. Difficulty adjusting Flow - Pump Interference Suspected
C. Run Aborted - Computer did not cycle
D. Flow meter upstream of filter for all subsequent Dow tests.

This approach would provide data as to whether the phenomenon was area sensitive. This was indeed the case and there are some later indications that the indicated mass gain of the filter may be a function of different filter manufacturing lots. The data regarding this, however, is inconclusive at this point. Tables 3-2 and 3-3 synopsize the data from the non-leaded tests. A significant observation is that the percentage deviation between the 2 inch and 143 mm sample is of the same order as the beta gauge (20 mm diameter sample) and the 143 mm sample. This phenomenon is currently attributed to a higher moisture absorption and absorption of high volatility organics by the larger filter. Inasmuch as excellent data had been obtained with leaded fuels, it was decided to discontinue the non-leaded fuels and switch to low-leaded fuels.

3.2.3 Tests on Low-Leaded (0.5 cc/gal TEL) Fuels

This series was also run using a 1971 Chevrolet with 350 CID engine. There was some attempt during this series to vary certain parameters surrounding the sampling process and determine their effect but little conclusion can be drawn from these attempts. Tables 3-4 and 3-5 present a synopsis of the low-leaded tests.

Here again the beta gauge agreed quite closely in several cases with the 2 inch diameter filter. It is interesting to note that as the test mode switched from the LA-4 cycle to steady-state operation (tests 26 and 27), the polarity of the percentage deviation between the two inch filter and the 143 mm filter reversed, causing the beta gauge to give

TABLE 3-2

Test No.	Test Mode	Pump Time (minutes)	IN Mass Gain (μg)	IN Total Flow (liters)	Dow 143 mm Gain (μg)	Dow 2 in (Gain (μg)	IN Rate (mg/min)	Dow 143 mm Rate (mg/min)	Dow 2 in Rate (mg/min)
16	LA-4(C.S.)	23	1049	2454	4050	--	6.05	22.01	--
17	LA-4(H.S.)	10	156	1208	1350	--	1.83	16.87	--
18	LA-4(H.S.)	10	33	736	1400	--	.64	7.61	--
19	LA-4(H.S.)	23	230	2075	1400	--	1.57	7.61	--
20	LA-4(H.S.)	23	197	2378	1400	200	1.17	7.61	1.32
21	Calif. Cyc.	60	359	4750	2200	600	1.08	5.79	1.92

TABLE 3-3

Test No.	% Dev. IN-143 min 143 min	% Dev. IN-2 inch 2 inch	% Dev. 2 inch-143 min 143 min	Remarks
16	-72.5	--	--	
17	-89.1	--	--	
18	-91.6	--	--	
19	-79.4	--	--	
20	-84.6	-11.3	-82.9	
21	-81.0	-43.8	-66.8	Test terminated at 47.5 min. (Automotive Failure)

NOTES:
1. Dow 143 mm flor - 4CFM
2. Dow 2 inch flow - 3.3 CFM
3. Fuel is non-leaded
4. Vehicle is 1971 Chevrolet - 350 CID engine
5. 2 inch filter had 6.5 in Hg differential pressure
6. 143 mm filter had negligible differential pressure

TABLE 3-4

Test No.	Test Mode	Pump Time (minutes)	IN Mass Gain (μg)	IN Total Flow (liters)	Dow 143 mm Gain (μg)	Dow 143 mm Flow (liters)	Dow 2 in Gain (μg)	Dow 2 in Flow (liters)	IN Rate (mg/min)
22	LA-4(C.S.)	23	2796	1784	1700	2600	8200	2140	22.2
23	LA-4(H.S.)	10	960	1222	1600	1130	900	931	11.1
24	LA-4(H.S.)	23	251	506	2300	2600	1300	2140	7.0
25	LA-4(H.S.)	23	298	506	2400	2600	1400	2140	8.3
26	LA-4(H.S.)	23	959	1840	2500	2600	1400	2140	7.4
27	60 mph	15	4119	993	7000	1400	600	1400	58.7
28	30 mph	15	350	996	1200	1400	400	1400	5.1
29	15 mph	15	332	1469	700	1400	300	1400	3.2
30	Idle	15	248	1488	500	1400	200	1400	2.4
31	30 mph	15	510	1472	1500	1700	500	1400	4.9
32	60 mph	5	2308	496	3000	566	1600	465	65.5
33	60 mph	5	1605	495	2200	566	1200	465	45.7

TABLE 3-5

Test No.	Rate (mg/min)	Rate (mg/min)	% Dev. IN - 143mm 143 mm	% Dev. IN - 2 in 2 in	% Dev. 2 in - 143 mm 143 mm	Remarks
22	38.6	54.2	-42.4	-59.0	+54.1	IN Flow/2in Flow not constant
23	20.0	13.7	-44.4	-18.7	-31.7	Flows steady
24	12.5	8.6	-43.9	-18.4	-31.3	Face velocities matched on IN and and 2 in
25	13.0	9.3	-36.3	-10.3	-29.1	Gelman Type A filter used
26	13.6	9.3	-45.7	-20.3	-31.9	
27	70.9	52.5	-17.2	+12.0	-26.0	143mm - 2 in flow rates matched
28	12.2	4.1	-58.0	+26.0	-66.7	143mm - 2 in flow rates matched
29	7.1	3.0	-55.0	+ 5.0	-57.1	143mm - 2 in flow rates matched
30	5.1	2.0	-53.4	+16.4	-60.0	143mm - 2 in flow rates matched
31	12.5	5.1	-60.8	- 3.2	-59.5	143mm flow returned to 4 cfm
32	75.0	48.6	-12.7	+34.6	-35.1	Doors to facility open
33	55.0	36.5	-16.8	+25.4	-33.7	Doors to facility closed

an indication which is between the two gravimetric measurements. There was a very evident change in moisture content between the two modes with some condensate appearing on the clear plastic tubing which was used during the La-4 cycle and no visible condensate during the steady state runs. The temperature of the sample flow did not appear to change appreciably on any of the runs besides the 60 mph steady-state runs. There is also a higher concentration of volatile organics during the La-4 cycle and this may also be a major contributing factor.

An experiment was also run by varying the temperature of the room in which the automobile was located. This was accomplished by running a test with the front and rear doors of the dynamometer facility open, then duplicating the test with the door nearest the front of the automobile closed. This produced a temperature variation of 30°F in the room and tests 32 and 33 are the results. Notice that there is little difference between tests 27 and 32, both being run with the doors open. Test 33, however, showed a significant reduction in particulate emission resulting from raising the intake air temperature to the engine.

3.3 Conclusions Regarding the Field Tests

The field tests at Dow and Cincinnati were sufficient to establish the capability of beta gauging as a process monitor. The detector stability achieved and the use of digital processing throughout assures high computational accuracy and repeatability. Much, however, remains

to be learned about the sampling process. The fact that poor correlation is achieved between filters of different diameters must surely be more fully explored and understood.

The data on moisture absorption by paper filters also indicates this medium to be undesirable for most applications requiring resolution of 1 - 2% of filter basis weight. Such materials as the GE Nuclepore or Gelman Type A do not exhibit as serious a problem in this area as the Pallflex E70/2075W and it is understood that these materials are under investigation by the EPA at the time of this writing. Other materials must be investigated and the filter industry must be called upon to address this problem.

Variations of a percentage of initial filter basis weight would affect both gravimetric and beta gauging identically. The larger filter face velocity of the beta gauge, however, would tend to concentrate the material and result in a larger basis weight change for the particles. For example, the effective sample area for the beta gauge is approximately 3 cm^2 while for the 2 inch gravimetric comparison, the effective area is approximately 10 cm^2. At 4.2 cfm for the beta gauge, this represented a normalized flow rate of 1.4 $ft^3/(min \cdot cm^2)$. At 3.3 cfm for the gravimetric unit, the normalized flow rate is 0.33 $ft^3/(min \cdot cm^2)$. Obviously, then, for the same stream concentration, more material will be collected on the beta gauge's filter on a per-square-centimeter basis and the moisture variance will not be as significant.

It is not the intent of this document to attribute the inconsistent data solely to moisture. Rather, this was an area in which IN has considerable expertise and facilities to investigate (at least under static conditions) from our previous experience in the paper industry. It is the intent, however, to point out to the reader that moisture can (and in many cases likely will) be a major cause of inconsistent data. The effects of filter composition on "errors" in data are relatively unknown to IN, and as a result, little positive or conclusive data can be presented in this area.

4.0 RECOMMENDATIONS

There are several areas of study and clarification revealed as a result of this program. First, it is believed that some definition is required by the Environmental Protection Agency as to what constitutes "particles". This may eventually require an exact preparation of the mixing tube air with respect to temperature and humidity. It may also require withdrawing a sample from the tube at a reasonably fixed flow rate (implying a constant pressure drop across the filter, barring any filter clogging).

In order to do the above, however, the problem of moisture interference must be solved. In IN's opinion, this is one of the major problems facing particle sample today. It should be of concern not only to those involved with mobile sources but also those concerned with stationary source or ambient monitoring. It is not sufficient simply to allow the gravimetric samples to stabilize in a controlled environment room as Figure 2-5 of this report will verify. In effect, in order to place any validity on data, the complete previous history of the filter must be either known or standardized and the nature of each sample must be known and taken into consideration. Solution may require either the discovery of a medium already produced but not widely touted or the manufacture of a new medium, using materials specifically designated to ward off water.

There is also limited data which shows that the composition of the filter medium may introduce wide fluctuations in the data. This is attributed to an affinity (or lack of same) for highly volatile constituents in the sample which are condensed from the stream and which would ordinarily remain in the gaseous state. A study should be initiated to determine the total nature of vehicular exhaust under both steady state and Federal Cycle operating conditions. This information could then be used to specify (or eliminate) certain materials used in the manufacture of the medium.

It is believed that extensive experimentation will be required by the EPA to establish correct and reliable sampling procedures. The proof of principle of beta gauging has been successful: a nucleonic gauge can be built with the required sensitivity. Further development in this area should be directed towards specific application areas rather than general purpose application. A filter medium must be found, not only for the beta gauge but for gravimetric comparison as well. The IN design is excellently suited to this type of experimentation by virtue of the cassette approach and should be able to yield valuable data in this area.

Finally, the Federal Cycle Test as currently understood by IN is at best cumbersome. It is believed that solution of the above two problems will yield far more consistent results than available in the past. It is also our belief that reliable data may also show that a

definite correlation exists between a typical steady-state sample (60 mph, for example) and the LA-4 Federal Cycle Test. If this be the case, a considerable reduction in the complexity (and therefore cost) of an automobile particulate emission gauge is possible.